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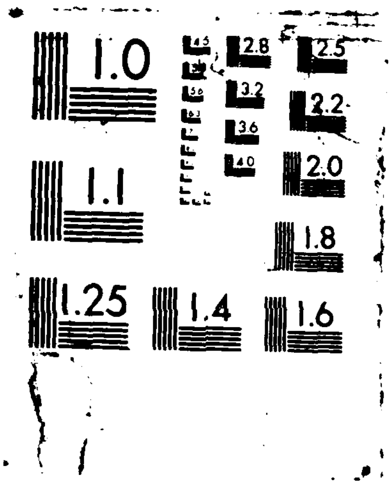
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Office of Naval Research

Final Report

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15 JUNE 1984 THROUGH 31 DECEMBER 1986

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"OPTICAL PROPERTIES OF HETEROSTRUCTURES AND SUPERLATTICES"

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Optical Properties of Heterostructures and Superlattices

I Introduction

The project for the study of the optical properties of single and multiple quantum wells involved both experimental and theoretical studies of such heterostructures.This study has utilized a variety of optical techniques such as laser excited luminescence, photo-reflectance, laser emission with and without magnetic fields.The initial experiments which were conducted on a single quantum well of GaAs/GaAlAs structure led to an interesting discovery and explanation of exciton transport at low excitation intensities at a critical range of temperatures. The second class of experiments involved high intensity laser excitation which led to the discovery of a new lower high density excitonic state below the lowest exciton when under moderate excitation. The identification of this new state was facilitated by the use of high magnetic fields. A corollary to this experiment was the observation of the electron-hole plasma state at the highest intensities .This occurs at still a lower energy and was shown to be the source of stimulated emission in a quantum well.

The most difficult aspect of the photoluminescence experiments involved the Zeeman studies of the excitons in QW structures. The experiments which were performed in a Faraday configuration using circularly polarized light to identify the Zeeman structure showed extra structure of both the light and heavy hole excitons.The complication of the study was the theoretical interpretation of the spectra..The QW clearly splits the light and heavy hole excitons at zero field and allows a two dimensional analysis which in effect separates the coordinates and the effect of the QW and the magnetic field. However the latter mixes the light and heavy hole states and the simple interpretation no longer applies. The initial studies have explained the phenomenon of the splitting semi-quantitatively. A more recent project using photoreflectance techniques has permitted higher field studies.

The theoretical analysis of this has also been refined in some respects. A paper combining these two studies is planned for the near future as a comprehensive publication of the Zeeman effect in a quantum well.

Another class of experiments involving electrically excited single quantum wells in lead chalcogenide lasers at high excitation levels has also been examined.These show spectra which involve forbidden transitions. These have been qualitatively explained by second order perturbation theory which shows mixing of the states in the quantum well and hence break down the selection rules. Magneto-optical spectra show even more complexity and are difficult to explain quantitatively. This work has been the subject of a graduate thesis. Although this project was terminated upon graduation it inspired another more recent study in GaAs/GaAlAs diode lasers in magnetic fields. The magneto-optical spectra of laser emission are less complicated and should be amenable to analysis since the theoretical basis for the Zeeman studies of excitons in QW in this case has been well established.

The last class of experiments that have evolved from those earlier studies involve reflectance, which complement the photoluminescence studies. The former have been made possible by the development of optical fiber methods which made magneto-reflectance possible. These techniques were developed under the Navy contract as part of a thesis project. The techniques have been used to study both single and double quantum well structures. With the former a detailed theoretical study of the line shape has been completed and accepted for publication. These studies again involve GaAlAs-GaAs structures. The magneto-reflectance studies have been carried out to higher fields. The theoretical interpretation of the higher order Zeeman effect has been quantitatively refined by the use of improved variational wave functions of the two dimensional analysis of the exciton in a QW. These recent studies and experiments on magneto-reflectance and photoreflectance are the subject of a current thesis.

Two theoretical studies were initiated, one on a periodic heterostructure laser, the other on the analysis of a CdTe-HgTe QW structure which was intended as an infrared detector. Both were discontinued since experimental programs could not be carried out to demonstrate the concepts. The one on the periodic structure would have required the resources and dedication of a group with an MBE machine committed to the development of such a device. Funds were not available. The second project on the IR detector was attempted experimentally with negative results.This was not inconsistent with the theoretical estimate which predicted a marginal effect, although not conclusive. The conclusion was that the project was not worth pursuing.

II. Exciton Transport in GaAs- GaAlAs SQW

The experiments on optically excited GaAs-GaAlAs single quantum wells (SQW) at low laser excitation levels as a function of temperature between 1.9 and 16 K exhibited some unique exciton transport properties. In a publication in J. Appl. Phys.55,4367 (1984) we discussed in full detail the nature of this phenomenon and its quantitative behavior. A copy is included with this report. However we will summarize the important results of this work here.

The experiment utilized a CW HeNe laser at 6828 Å to create electron hole pairs throughout the entire structure. Thus most of the pairs created occupied the GaAlAs barrier region. These pairs were then free to travel to the GaAs QW region. The experiments definitively established that the pairs formed excitons and travelled as neutral pairs.The GaAs QW acted as a sink for the excitons which then recombined at the lower energies associated with the QW. The exciton luminescence in the barrier region decreased with increasing temperature. High magnetic fields had no effect on the process, indicating that the excitonic transport rather than free carriers were involved. Correlation between theory and experiment is presented in the enclosed reprint.

III. High-Density Excitonic States in 2D MQW

The second experiment utilized a high intensity dye laser pumped with a doubled YAG laser.The dye laser was tuned to excite electron hole pairs just above the heavy hole exciton energy in a multiple quantum well (MQW) of a GaAs-GaAlAs structure.The intensity of excitation was varied from a few W/cm^2 to as high as 1MW/cm^2 .At low excitation intensities the spectral structure was similar to those observed with CW excitation of a HeNe laser.As the intensities increased the heavy hole line broadened and at intensities above 10kW/cm^2 a new relatively narrow structure appeared below the heavy hole exciton and persisted distinctly up to about 100kW/cm^2. Above that level the electron-hole plasma (EHP) line began to appear at a lower energy and at about 1MW/cm^2 was the dominant feature.

The interpretation of this new feature which persisted for the intermediate high excitation intensities at energies between the lowest excitonic level and the EHP was that it is a collective excitonic state. That there was no sudden Mott transition from the solitary exciton to the plasma state, but instead the appearance of an intermediate excitonic structure was supported by three important observations. First the line was relatively narrow, of the order of 2 meV,comparable to the heavy hole exciton (HHE) line. In addition the Zeeman effect was quadratic in nature and also very similar to that of the heavy hole exciton.The last observation was that the relative energy of the structure to that of the HHE was dependent on the width of the quantum well.For further details see the enclosed reprint, Phys Rev. B 32, 1419 (1985).

IV. EHP in GaAs-GaAlAs MQW

One of the byproducts of the high intensity excitation studies of the multiple quantum wells (MQW) in GaAs -GaAlAs structures was the observation of the electron-hole plasma (EHP) line luminescence. This occurred at energies below that of the HHE and that of new high density excitonic state. The carrier densities associated with the EHP

varied from 10^{11}- $10^{13}/cm^3$, representing densities comparable to those attained in semiconducting lasers at threshold. Indeed when the gain measurements were made in the QW along its length, there was a definite indication of stimulated emission. The analysis of the luminescence of the EHP showed an energy gap renormalization of about 2 excitonic rydbergs. The lineshape analysis was consistent with a 2D density of states. The excitation near the HHE level produced a surprisingly low plasma temperature of the order of 25 K, but not necessarily inconsistent with the relatively low energy of excitation i. e. 10 meV difference between the laser energy and that of the plasma peak. Finally the magneto-optical studies of the EHP revealed a quenching phenomenon with increased magnetic field. This is most evident for low density EHP, when the electron cyclotron energy is comparable to the Fermi energy.

The actual results of the calculations and experiments are shown in the enclosed figures. Fig. 1(a) shows the dispersion relation for the conduction and valence bands in a quantum well. Fig. 1(b) shows the E-k contours of the lowest hole band and Fig. 1(c) shows the occupation function of both electrons and holes , which have been calculated as a product of the density of states and the Fermi-Dirac distribution. Fig 2(a) shows the theoretical calculations of the recombination lineshape for different values of electron-hole densities. Fig 2(b) shows the experimental results of the EHP luminescence for different levels of excitation intensities. Both theory and experiment exhibit the consequences of a 2D density of states. Fig. 3 shows the results of the experiments and the theory for the effective ground state energy of the EHP as a function of $r_s \sim N^{-1/2}$, where N is the plasma density. E'_G is normalized by the effective rydberg R* of the exciton. The comparison between experiment and theory, as indicated by the open circles and the solid line, is quite satisfactory. These results were reported at the APS meeting, see Bull. Am. Phys. Soc. 30, 382 (1985).

V. Zeeman Effect in Quantum Wells

The study of magneto-optical effect by luminescence techniques essentially is the study of the Zeeman effect of excitons in quantum wells. The experiments were carried out in a number of SQW heterostructures of GaAs-GaAlAs samples with QW widths varying from 100 Å to 175 Å. The samples were mounted in a dewar and in a Bitter magnet in fields up to 100 kG. Excitation was performed with a HeNe laser in a Faraday configuration. Luminescence was analyzed with a spectrometer collecting light at right angles to the QW and detecting it with a GaAs cathode photomultiplier tube. Spectra were taken at temperatures below 2K and excitation intensities less than 0.5 W/cm^2.

Luminescence spectra with L = 175 Å at zero field are shown in Fig. 4(a) These are the degenerate exciton doublet which are labeled X_h and X_l light and heavy hole excitons which are split by the QW. In a magnetic field each in turn shows a Zeeman splitting which is best observed with circularly polarized light, labeled σ_+ and σ_-. The structure or splitting is shown in Fig.4(b). In addition we show the excited at higher energies in Fig. 5 These are then plotted as a function of magnetic field in a fan chart shown in Fig.6. The diagram shows a quadratic shift as well as the spin splitting of the ground states of X_h and X_l. The solid and dotted curves are theoretical and show an excellent fit to the experimental data.

The analysis of the Zeeman structure is a complicated one which is the subject of a current thesis and will be published in the near future To analyze the Zeeman effect of the exciton in a QW we set up a Hamiltonian which consists of a 4x4 matrix representing the holes and electrons. The latter appear as the diagonal terms together with the QW potential. The magnetic terms appear in both the diagonal and the off-diagonal components. The latter are treated as a perturbation and hence the problem is reduced to the solution of four individual equations with reduced mass parameters. The problem is then solved by introducing a 2D Coulomb term $\lambda e^2/\rho$ with a variational parameter λ first introduced by Jiang. This reduces the whole problem, after separation of variables, to a 2D excitonic problem in a magnetic field The QW solution appears in the z coordinate as a separate

equation. The hydrogen problem in the ρ coordinate is solved by a variational technique for a particular value of λ, which is magnetic field dependent.In the Faraday configuration the QW solution is magnetic field independent. Once the 2D solution for the exciton is obtained in the diagonal component form, the resultant wave functions are used to second order to obtain the correction due to the off-diagonal components. These couple the heavy and light exciton states and properly account for the Zeeman structure of the excitons shown in Fig.4. This analysis including a more precise treatment of the ground state quadratic Zeeman effect at higher magnetic fields will be described in the thesis by Xiao-Lu Zheng entitled "Magneto-optical Properties of GaAs-GaAlAs QW Structures".

VI. Quantum Well Lasers

One of the interesting phenomena examined in the study of quantum wells was the stimulated emission of QW lasers with and without a magnetic field. The particular lasers that showed the most unique results were those of the lead salt compounds. When these were studied as a function of increased current in a diode structure above threshold, they exhibited an unexpected multiplicity of emission lines which seemed to violate the normal selection rules for QW transitions, i.e. $\Delta n=0$. The answer to this apparent dilemma resides in the fact that the energy momentum relations of the energy bands in the lead salts are represented by ellipsoids along the [111] axes. The QW is however grown along the [100] axis. Hence in order to solve the energy level problem in the QW we first rotate the ellipsoidal functions to the crystal coordinates or the [100] axes. When we solve for the energy levels to first order, the mixed momentum terms are neglected, i.e. $p_x \neq 0$, but $p_y = p_z = 0$.This is valid near the threshold. However when the laser is excited at higher intensities the mixed terms must be taken into account, since $p_y = p_z = p_F$ We do this by treating the mixed terms as a perturbation to second order.The result is that the states with quantum numbers n = 0, n= 1 and n =1, n =2 ,etc. are mixed. This leads to allowing transitions with selection rules $\Delta n = +1$ and $\Delta n = -1$ and hence we can account for the

observation of the additional lines. The details of the experiments and the theory are presented in the thesis by Gary Lorenzen entitled " Behavior of $Pb_{1-x}Sn_xTe$ Semiconductor Diode Lasers in a Magnetic Field", May 1984.

VII. Photoreflectance and Magneto-Reflectance in GaAs-GaAlAs QW Structures

In order to extend the magneto- optical studies to higher fields where the optical access to Bitter solenoids is limited, the technique of photoreflectance using fibers was developed. However in order to interpret the spectra properly the initial study was directed to examining the nature of photoreflectance line shapes. The apparatus to perform such experiments is shown in Fig.7. The components are standard items arranged as shown and appropriately labeled. Both reflectance and photo reflectance can be obtained simultaneously, The sample holder which is connected to three optical fibers can be immersed in either liquid helium or nitrogen and also can be placed in the bore of a Bitter magnet.

Typical photoreflectance lineshapes at different temperatures are shown in Fig.8. Magneto-reflectance and photoreflectance (PR) are shown in Fig.9. The PR as indicated by ΔR is much more sensitive and shows structure which permits the investigation to higher energies. The results of the PR spectrum as shown in Fig.10 extend from 1520 meV TO 1660 meV, an energy span twice that of the luminescence data of H. Le et al. In addition the magnetic fields have been extended to 15T as compared to 10T in the previous experiments.

To properly analyze the reflectance lineshapes we studied a variety of samples which had the single QW located at different positions relative to the incident surface where the reflection occurred. We observed that the lineshapes changed as a function of position or the thickness of the barrier layer which separated the quantum well and the surface. The reflectance was analyzed in terms of an interference phenomenon and the complex index of refraction or dielectric constant associated with the exciton absorption and dispersion in the

QW. Calculations of the lineshapes show good agreement with the observations. In addition we determine the oscillator strength of the exciton in the process of this analysis. The further benefit is that this study also provides the basis for analyzing the photoreflectance lineshapes as well. A description of the data and the theory is given in the paper in Appl. Phys. Lett. 52 287 (1988).

One final result we want to discuss is the definitive evidence of the observation of room temperature exciton in epitaxial GaAs QW structures by the use of magneto photoreflectance. The lineshapes in both bulk and QW structures are similar. In addition the lowest level shows a characteristic quadratic Zeeman effect associated with the exciton in both cases. Analysis of this is the subject of a refined 2D model of the ground state of the exciton in a magnetic field. The results of this experiment are discussed in the paper to appear in Appl. Phys. Lett. March 21 issue, 1988. A preprint is enclosed.

We have also used the photoreflectance and photoluminescence excitation techniques to study the excitonic effects in double quantum wells. these results together with theoretical analysis have clarified the conditons under which a doublet excitonic spectral structure is observable in the ground and the excited states.

VIII. Conclusion

The investigation of the optical and magneto-optical properties of QW structures under the ONR contract which spanned the years of 1984-86 has been very fruitful. The scope of the work which included the development of new experimental techniques, observation of a variety of new phenomena, accompanied by extensive theoretical analysis contributed to a greater understanding of excitons and plasmas in QW structures. The program established the combined techniques of laser spectroscopy and high magnetic fields as a powerful tool for the unravelling of the complexities of the phenomena observed, using these with quantum wells to measure the band parameters of holes and electrons with greater unambiguity and precision. The results are important in

understanding the application of the QW structures to such devices a QW lasers, non-linear switches, tunneling and other transport devices using such structures.

IX. Publications

1 - Le, H. Q., Lax, B., Maki , P. A., Palmateer, S. C., and Eastman, L. F., J. Appl. Phys. 55, 4367 (1984)

2 - Le, H. Q., Lax, B. Maki, P. A., Palmateer, S. C., and Eastman, L. F., Bull. Am. Phys. Soc. 29, 257 (1984).

3 - Lax, B. ,Le, H. Q., Maki, P. A., Palmateer, S. C., and Eastman, L. F. Bull. Am. Phys. Soc. 29, 256 (1984).

4 - Lax, B. ,Le, H.Q., Bull. Am. Phys. Soc. 30, 382 (1985).

5 - Le, H. Q., Lax, B. , Vojak, B. A., Calawa, A. R., Phys. Rev. B, 32, 1419 (1985)

6- Le, H.Q., Lax, B., Vojak, B. A., Calawa, A. R. and Goodhue, W. D., Proc of the 17th Intl. Conf. on the Physics of Semiconductors (Springer-Verlag, New York) 1985, p. 515-518.

7 - Zheng, X. L., Heiman, D., Lax, B. and Chambers, F.A. , Bull. Phys. Soc. 32, 872 (1987)

8 - Zheng, X. L., Heiman, D., Lax, B. and Chambers, F. A., Appl. Phys. Lett., 52, 287 (1988)

9 - Zheng, X. L., Heiman, D., Lax, B., Chambers, F. A. and Stair, K. A. Appl. Phys. Lett 52, March 21 issue,(1988)

10 - Zheng, X. L.,Heiman, D., Lax, B. and Chambers, F. A. Proc. 3rd Intl. Conf. on Superlattices, Microstructures & Microdevices, Aug. 1987, Chicago (to be published)

X. Theses

1 - Gary Lee, Lorenzen, " The Behavior of $Pb_{1-x}Sn_xTe$ Semiconductor Laser Diodes in a Magnetic Field ", May 1984.

2 - Xiao-Lu Zheng, " Magneto-optical Properties of GaAs-GaAlAs Quantum Well Structures " (in preparation for June 1988)

XI.Captions

Figure 1 (a) Dispertion relations for the conduction and valence bands in a quantum well. The labels +3/2, -3/2, +1/2, -1/2 are the spin quantum numbers of hole states quantized with respect to the QW axis. The mixing of these states causes non-parabolic dispersion.

(b) E-k countour of the lowest hole band. The unit of energy is the effective Rydberg (R^*), and the unit k-vector, inverse of the effective mass donor Bohr radius, a_B^*.

(c) Occupation functions of electrons and holes, which are products of-the-density of state function and the Fermi- Dirac distribution. At finite temperature and non-perfect crystal, the occupation functions deviate from the rectangular shapes shown in the dashed curves.

Figure 2 (a) Lineshape of EHP radiative recombination recalculated accoding to non-interacting single particle model, for various electron-hole pair densities. Plasma temperature is assumed to be about 20 K. Renormalized gap effect is not included.

(b) Experimental results on EHP luminescence, for various excitation intensities, I_{ex}. The dashed curve is the stimulated emission spectrum I_{ex} = 0.5 MW/cm^2 which is much sharper than the luminescence spectrum at the same I_{ex}. Both theory and experiment exhibit EHP lineshape with flat top, as a result of 2D density of states, which is different from the 3D case.

Figure 3 Average ground state energy per electron-hole pair, E_G, the chemical potential at $T = 0$, μ,the correlation energy ε_{corr},the renormalized band gap E'_g for 2D EHP are shown vs r_s defined by $N(\pi r^2_s) = 1$ where N is the electron-hole pair density. The solid curves are theoretical calculations. The open circles are the renormalized band gap determined from lineshape analysis of the experimental results.

Figure 4(a) The luminescence spectra of a single quantum well at zero field. X_h and X_l are the degenerate exciton doublet , the heavy and light hole excitons respectively, which are split by the QW potential.

4 (b) The Zeeman splitting of the exciton in a magnetic field as indicated by the solid and dotted curves obtained, with cicularly polarized light labeled σ^+ and σ^-.

Figure 5 The Zeeman spectra of the higher energy in states of the exciton in a QW as observed by photo-luminescence.

Figure 6 Fan chart of the Zeeman spectra showing the energies of the light and heavy hole exciton ground and higher states. The triangles and circles are experimental data and the solid and dotted curves are theoreical.

Figure 7 Schematic of the experimental apparatus used for reflecctance and photo-reflectance measurements of spectra in QW structures with and without magnetic fields. The latter facilitated by the use of optical fibers as shown.

Figure 8 Typical photo-reflectance curves vs energy for different temperatures in a GaAs-GaAlAs MQW

Figure 9 Magneto-reflectance and photo-reflectance spectra in a GaAs-GaAlAs MQW.

Figure 10 Fan chart of magneto-reflectance spectra in GaAS-GaAlAs MQW to higher energies and magnetic fields than those obtained by photo-luminescence in Fig. 5.

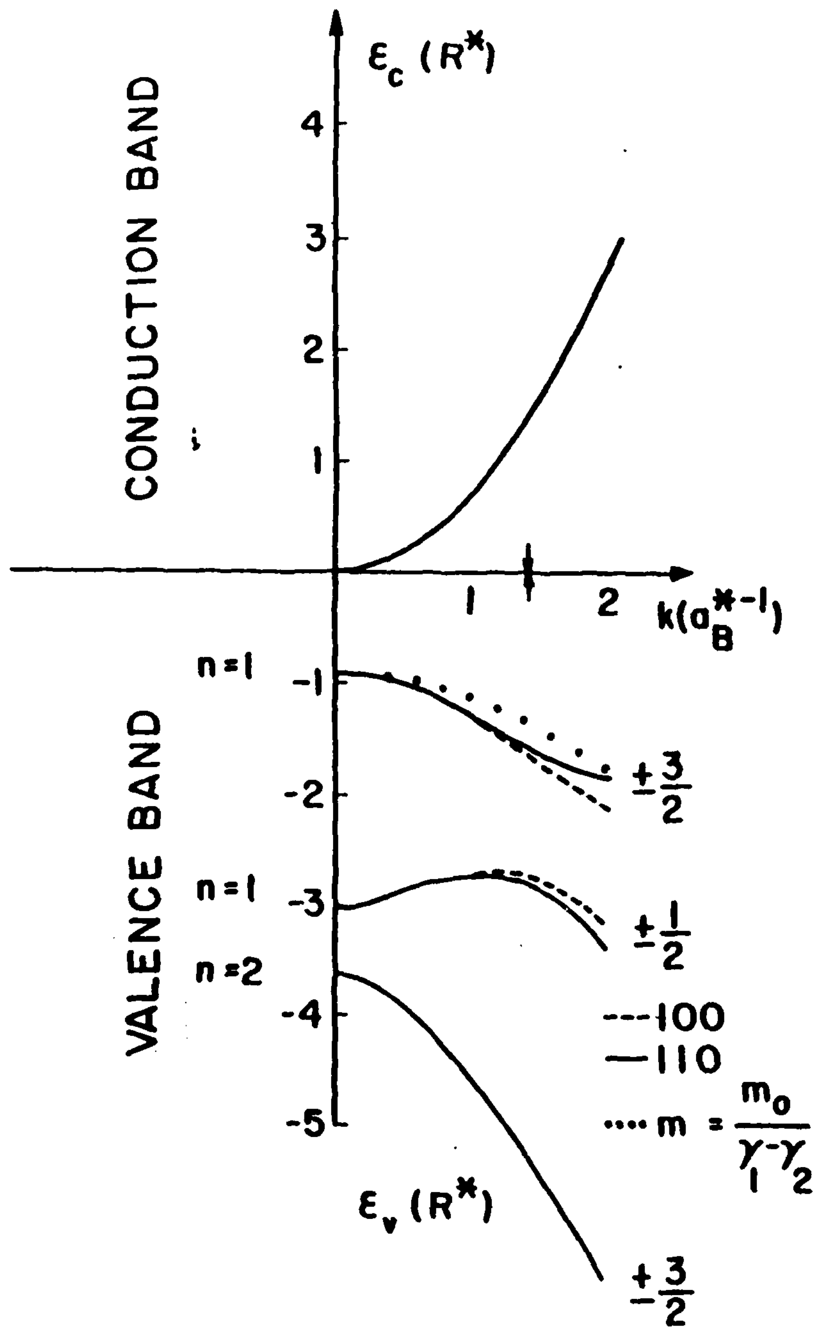

Fig 1 (a) Han Le

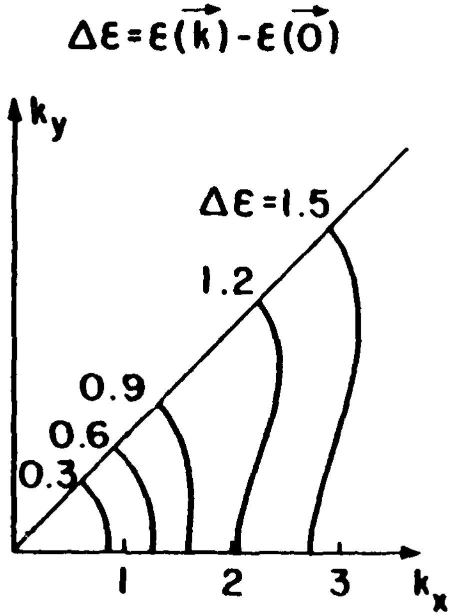

Fig 1 (b)

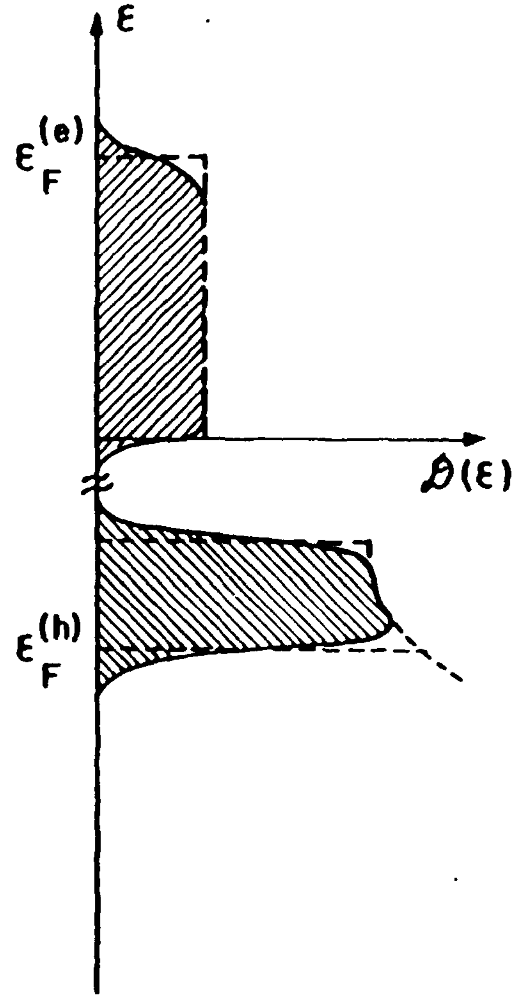

Fig 1 (c)

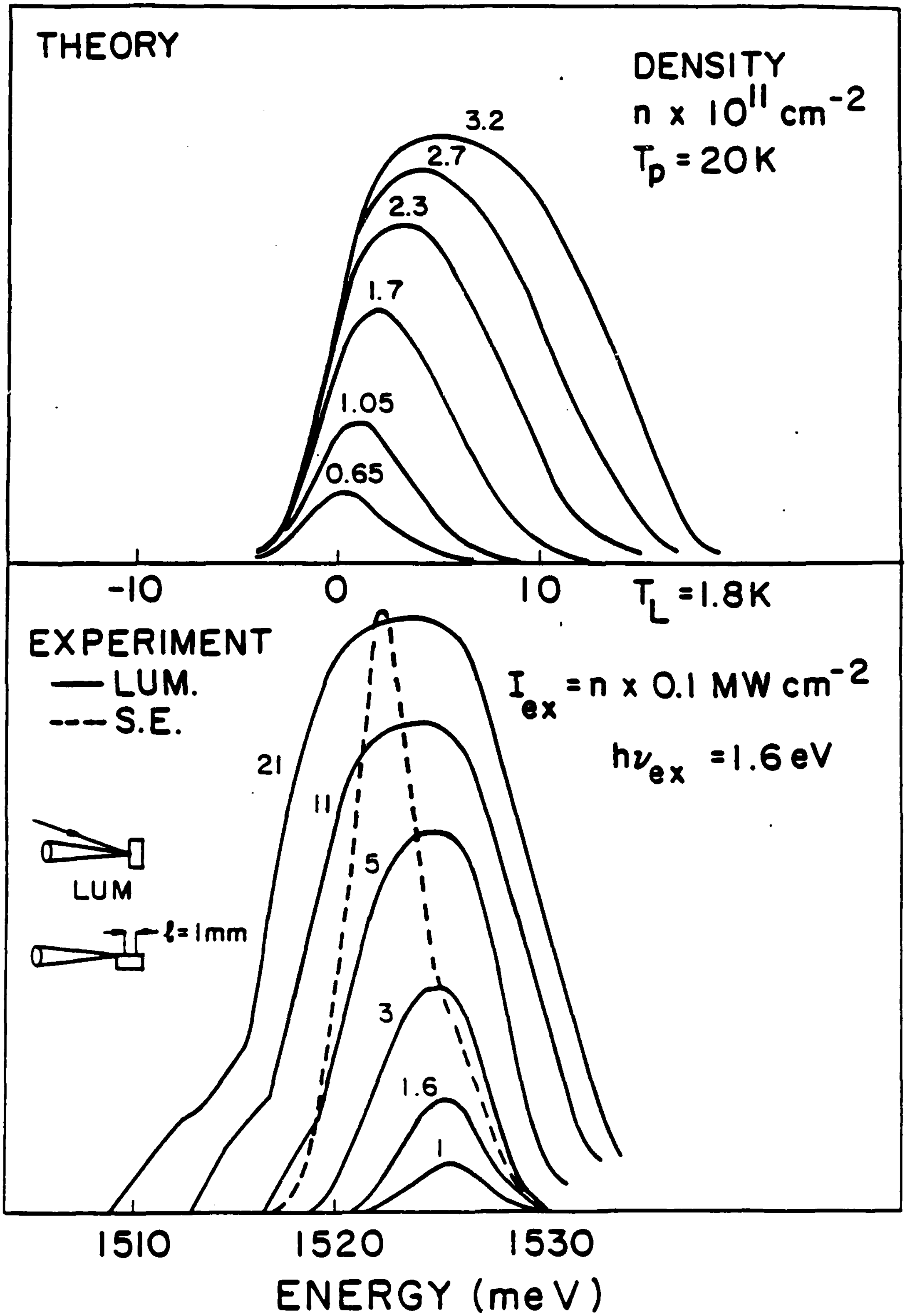
THEORY
DENSITY
n x 10^11 cm^-2
T_p = 20K
3.2
2.7
2.3
1.7
1.05
0.65
-10
0
10
T_L = 1.8K
EXPERIMENT
LUM.
S.E.
I_ex = n x 0.1 MW cm^-2
hν_ex = 1.6 eV
21
11
5
3
1.6
1
LUM
ℓ = 1mm
1510
1520
1530
ENERGY (meV)

Fig 2 (b) Han Le

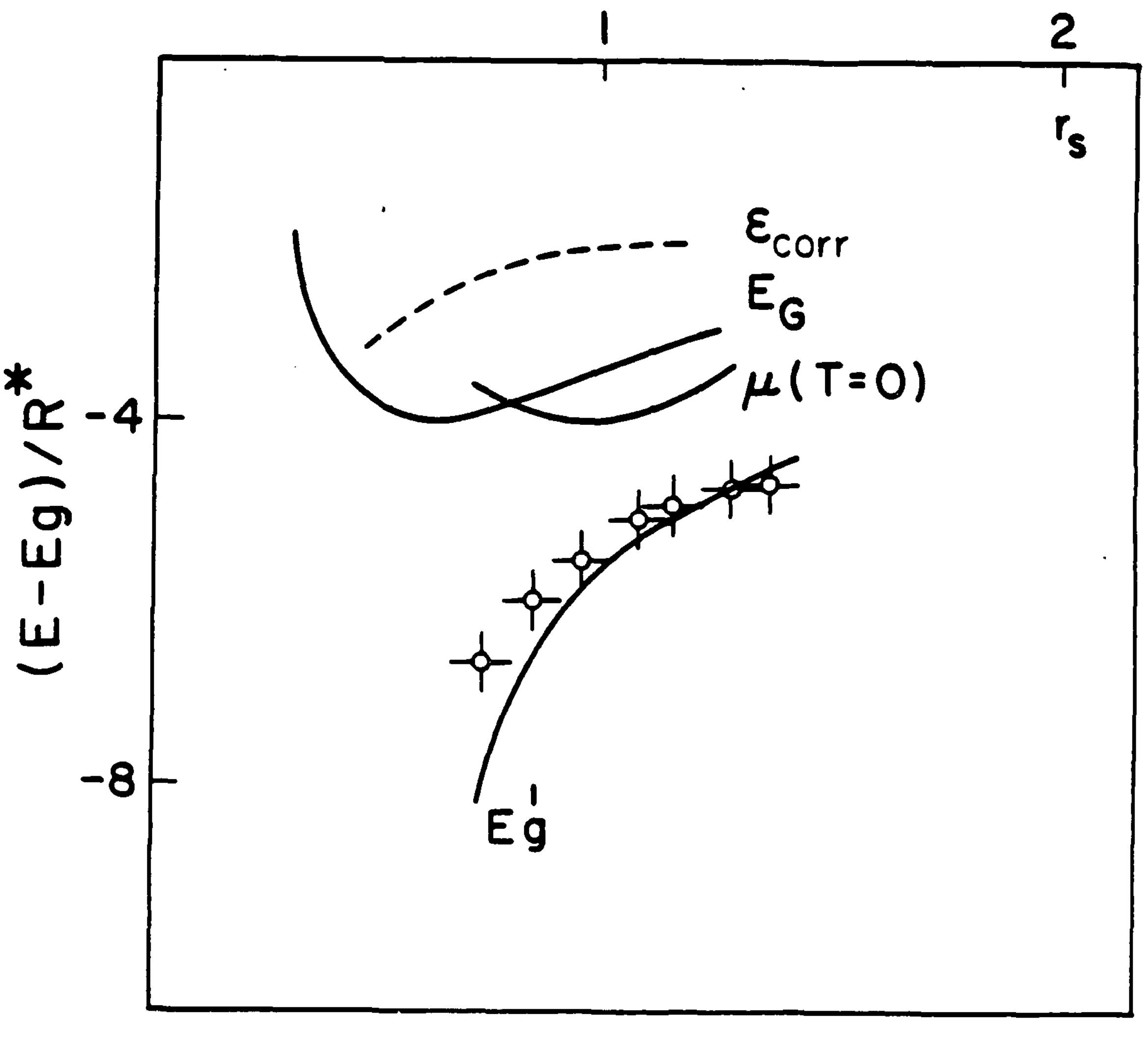

Fig. 3 Han Le

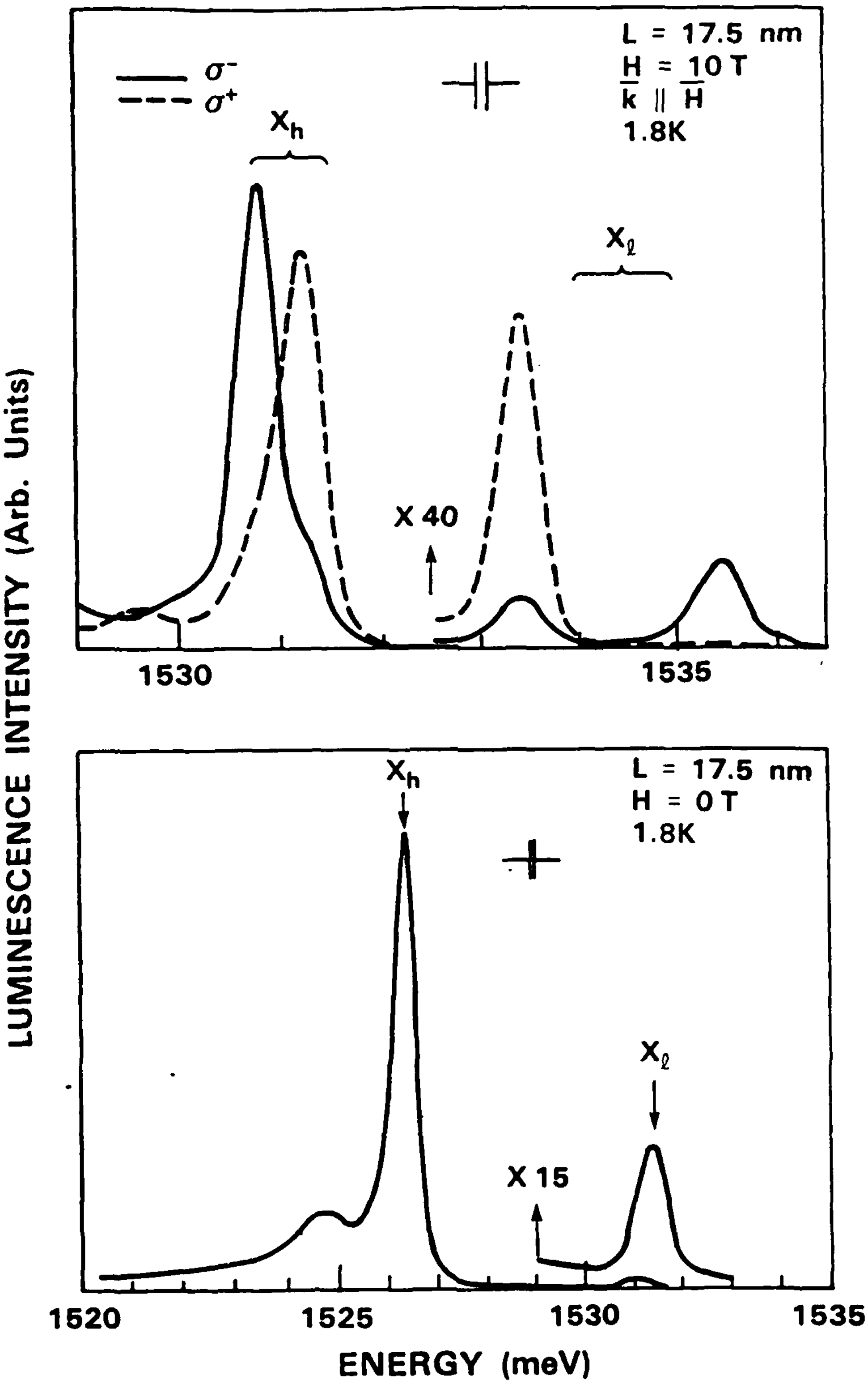

FIG 4(b)

FIG. 4(a)

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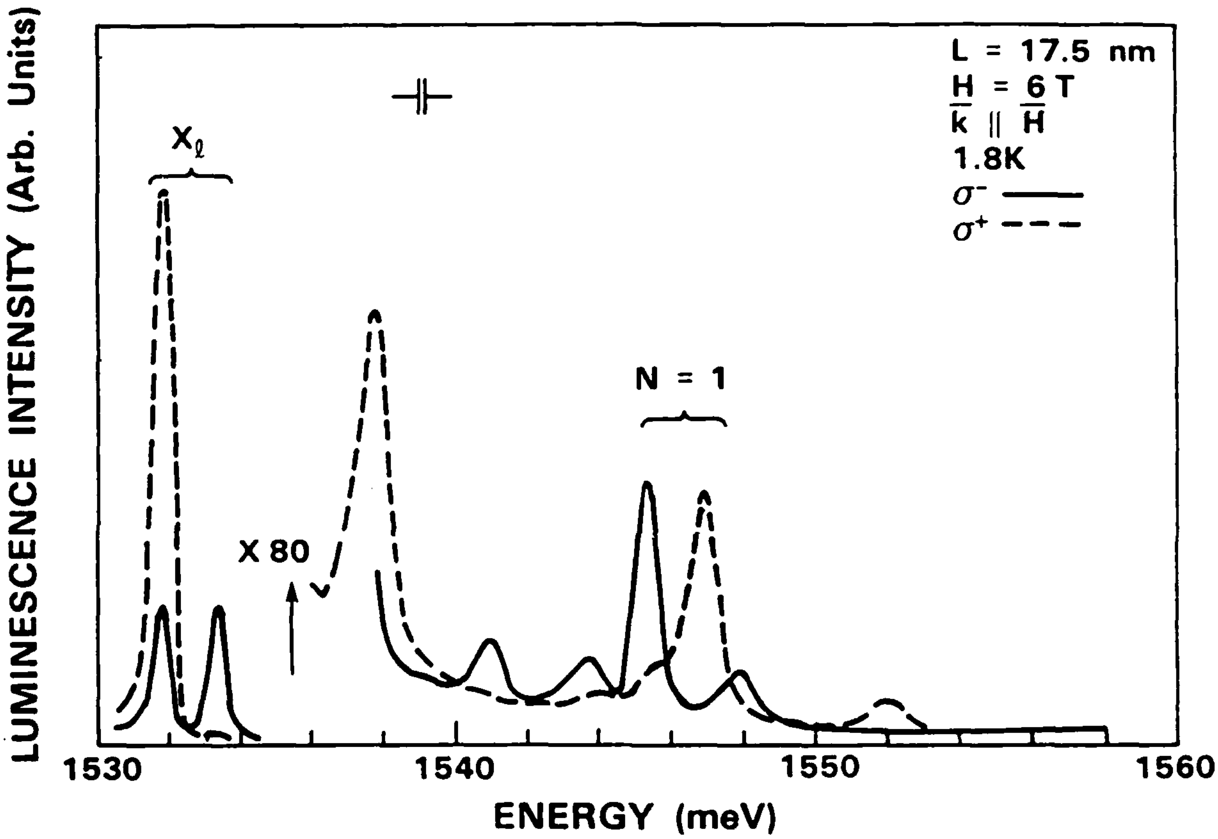
LUMINESCENCE INTENSITY (Arb. Units)
ENERGY (meV)
1530
1540
1550
1560
L = 17.5 nm
H = 6 T
k ∥ H
1.8K
σ^-
σ^+
X_ℓ
X 80
N = 1

FIG. 5

96153-1

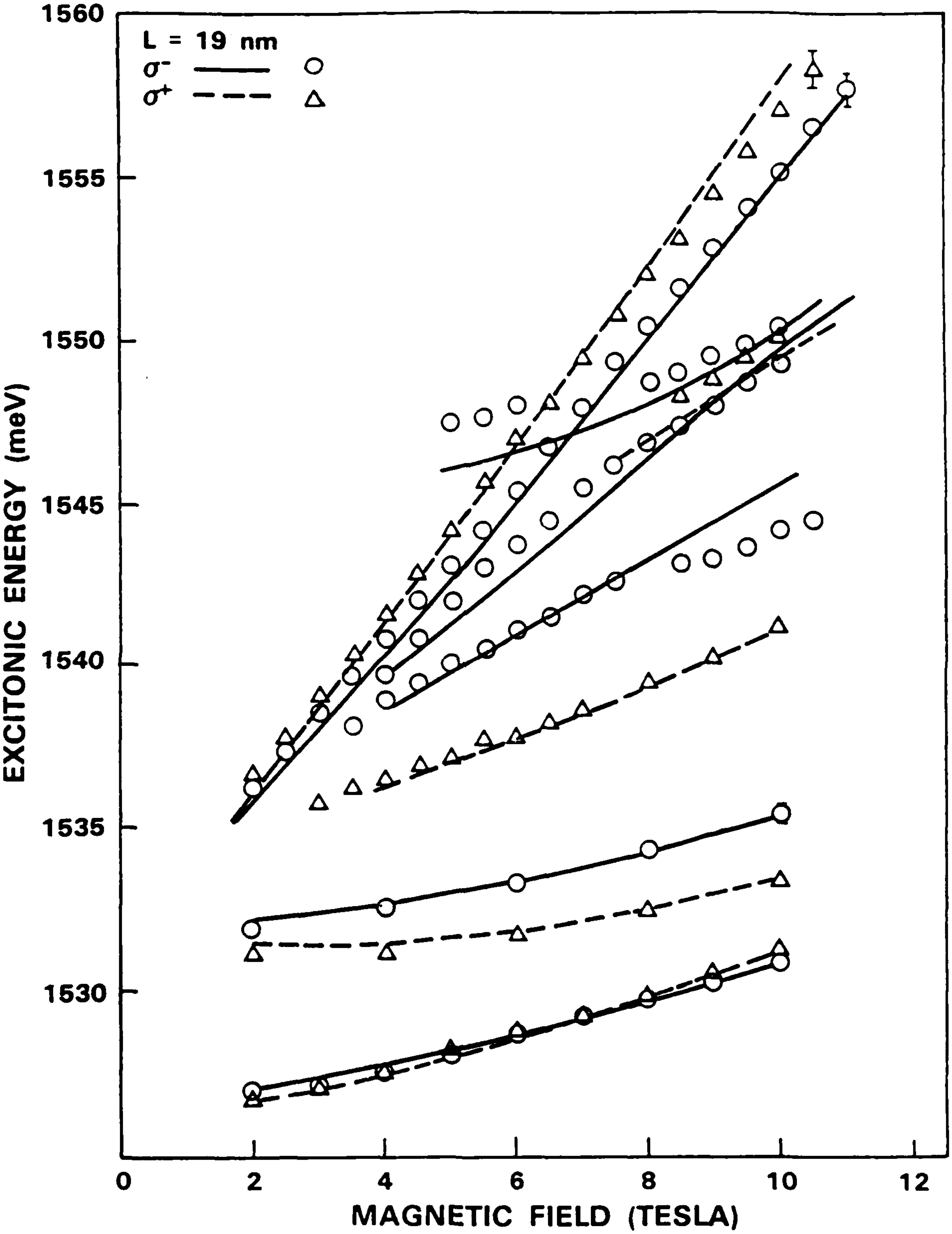
L = 19 nm
σ⁻
σ⁺
1560
1555
1550
1545
1540
1535
1530
EXCITONIC ENERGY (meV)
0
2
4
6
8
10
12
MAGNETIC FIELD (TESLA)

FIG. 6

96153-3

REFLECTANCE AND PHOTOREFLECTANCE SYSTEM

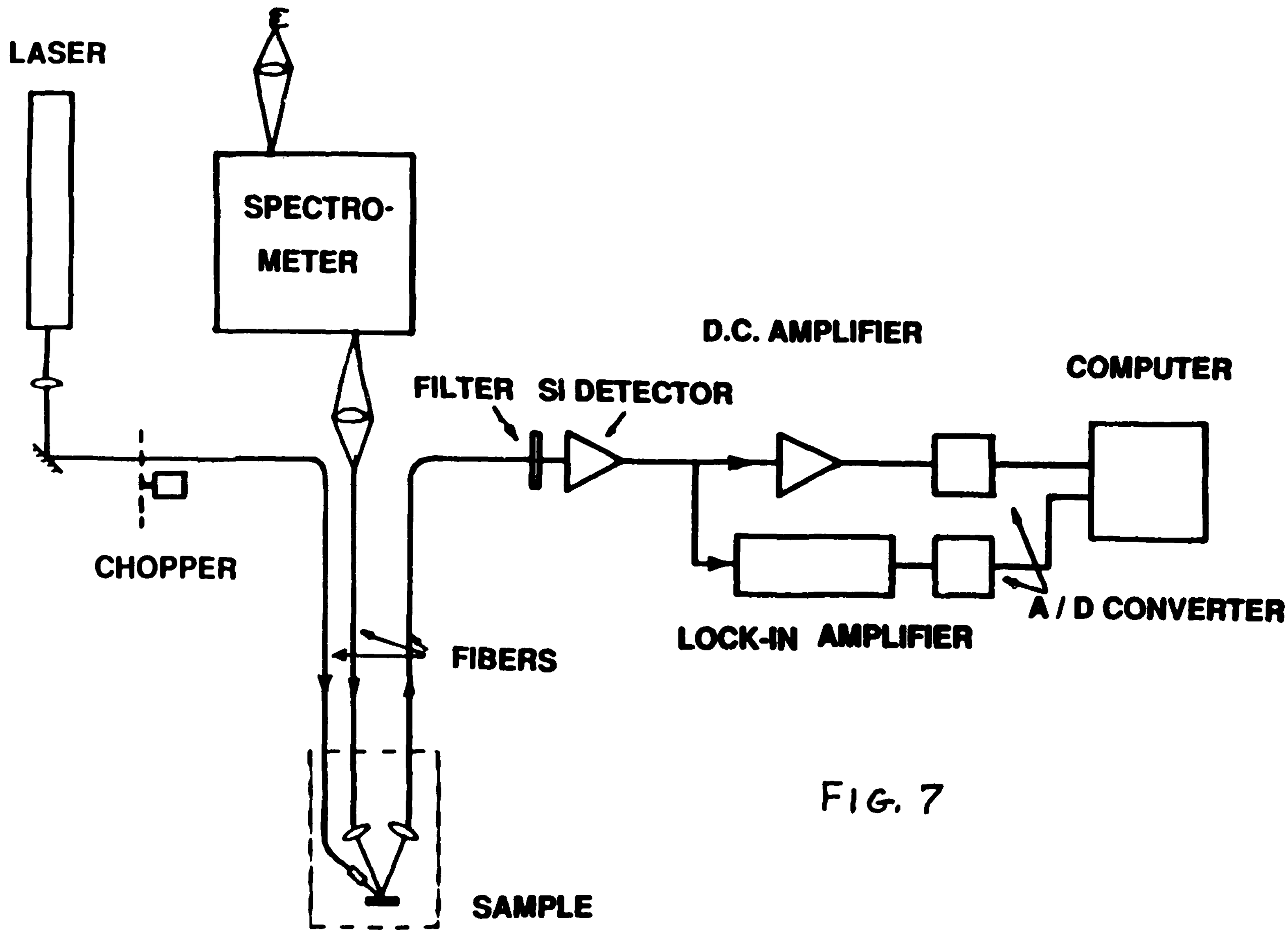

FIG. 7

TEMPERATURE DEPENDENCE OF PR

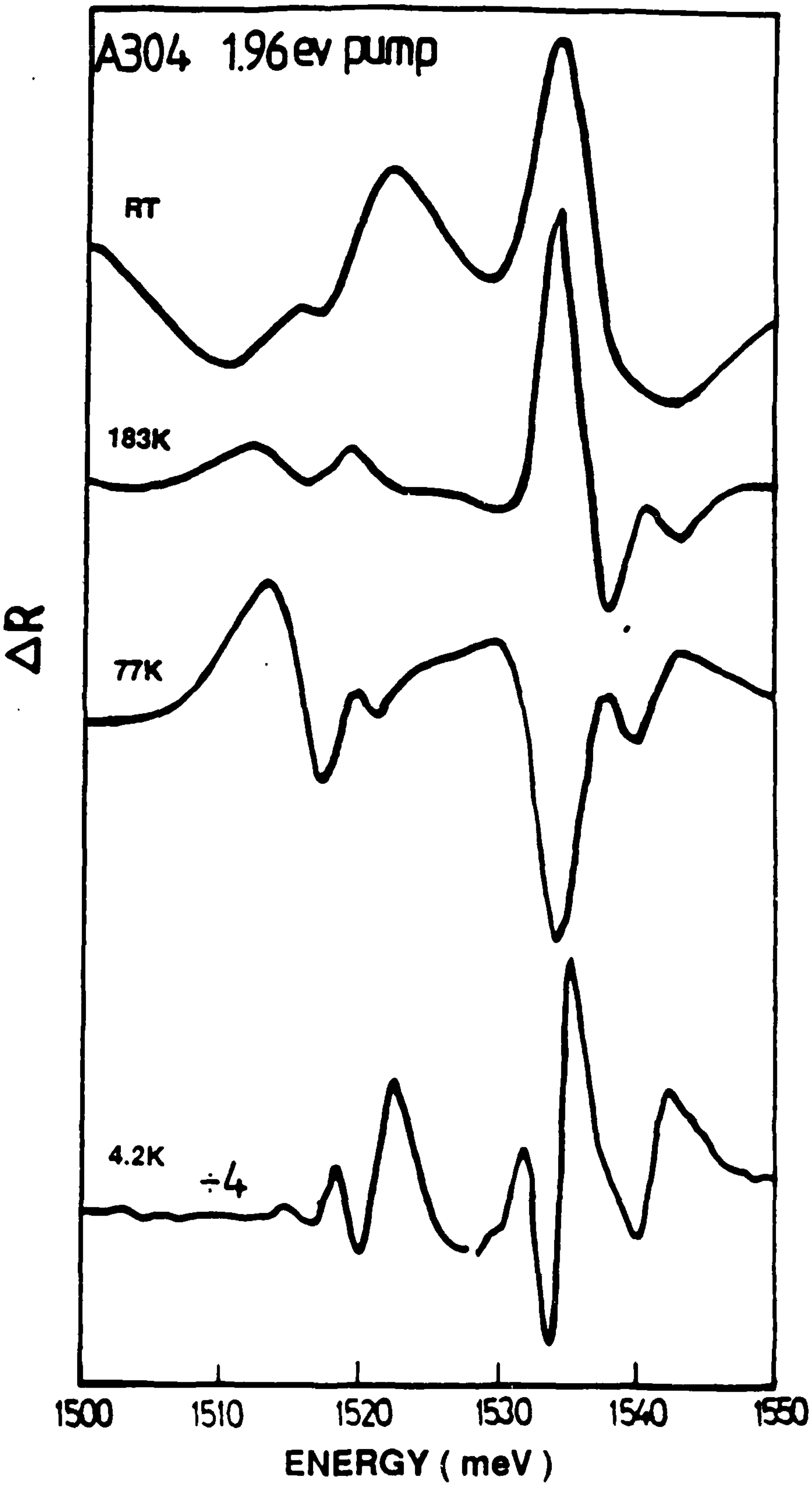

FIG. 8

SPECTRA OF MAGNETO-REFLECTANCE AND PR

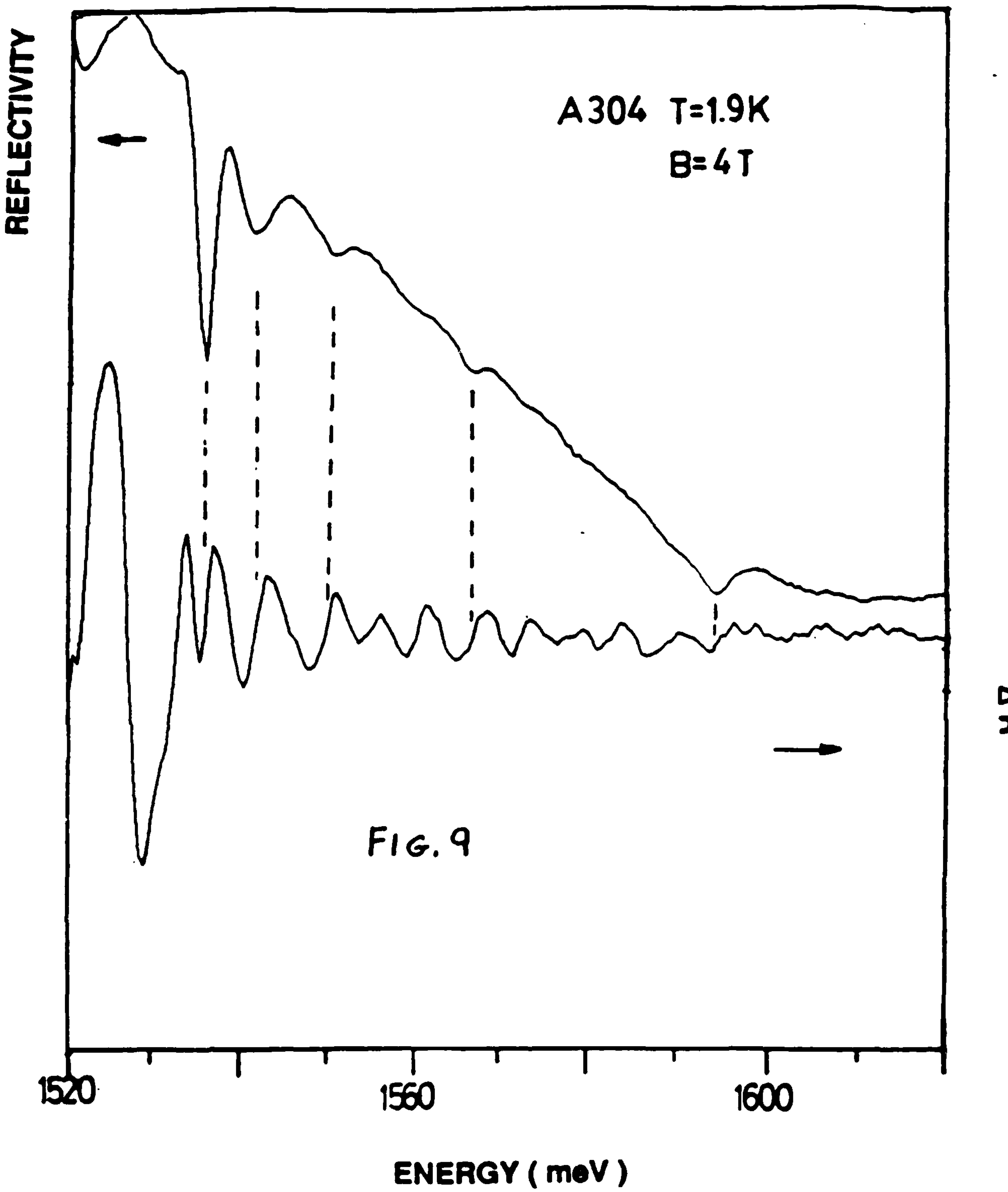

FIG. 9

INTERBAND TRANSITION ENERGY AS THE FUNCTION OF MAGNETIC FIELD

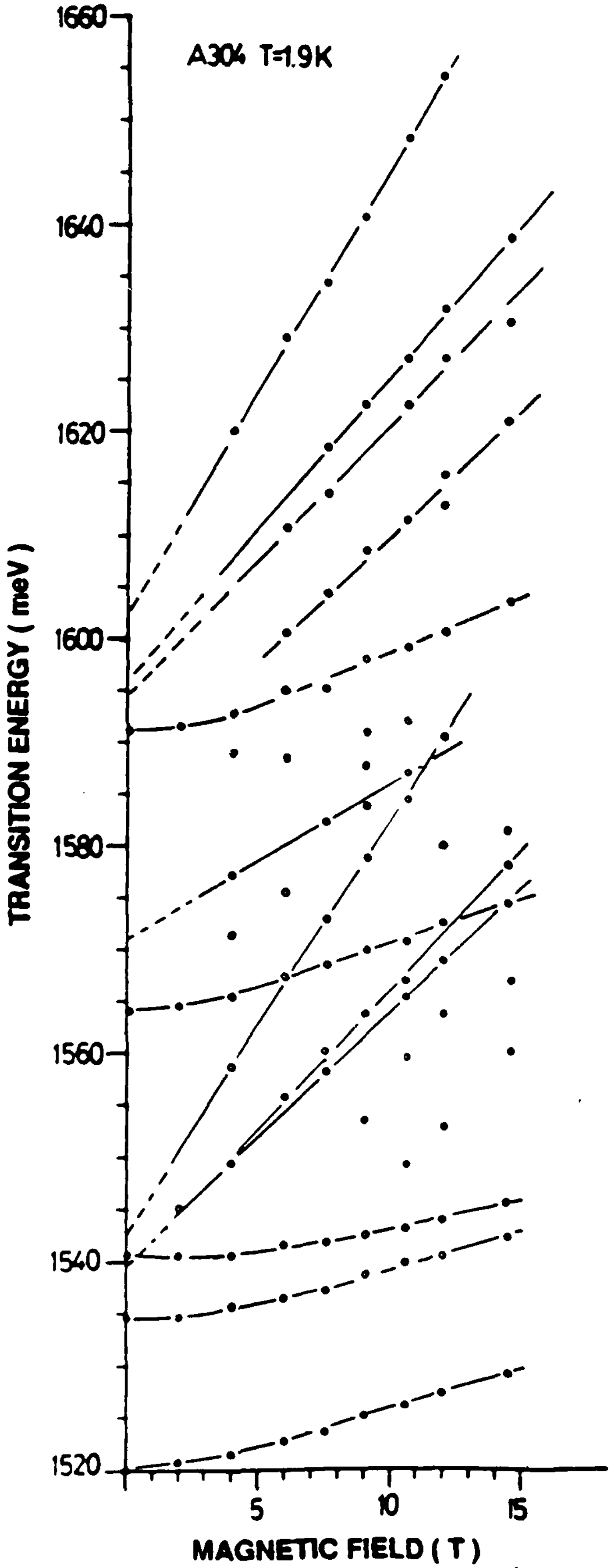

FIG. 10

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